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Evolution of two-gap behavior of the superconductor FeSe_{1-x}

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The superfluid density, ρ_s , of the iron chalcogenide superconductor, FeSe_{1-x} , was studied as a function of pressure by means of muon-spin rotation. The zero-temperature value of ρ_s increases with increasing transition temperature T_c (increasing pressure) following the tendency observed for various Fe-based and cuprate superconductors. The analysis of $\rho_s(T)$ within the two-gap scheme reveals that the effect on both, T_c and $\rho_s(0)$, is entirely determined by the band(s) where the large superconducting gap develops, while the band(s) with the small gap become practically unaffected.

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Since the discovery of Fe-based high-temperature superconductors (HTS) much effort is devoted to the investigation of their superconducting mechanism. While some properties of Fe-based HTS are reminiscent of the cuprate HTS (as, *e.g.*, their layered structure, the proximity to a magnetic phase, the universal “Uemura” scaling between the superfluid density, ρ_s , and the transition temperature, T_c), the differences between both compounds families are much more remarkable. Hence, the superconductivity in Fe-based HTS originates within the d -orbitals of the Fe ion, which are normally expected to lead to pair-breaking effects [1]. For the Fe-based HTS, several disconnected Fermi-surface sheets contribute to superconductivity, as revealed by angle-resolved photoemission spectroscopy [2, 3]. Furthermore, indications for multi-gap superconductivity was obtained from tunneling [4, 5], magnetic torque [6], point contact [7] and infrared spectroscopy [8] experiments, as well as from specific heat [9], first and second critical field [10, 11], and superfluid density [12, 13, 14, 15] studies. The multi-gap superconducting state positions the Fe-based HTS together with MgB_2 – the most famous double-gap superconductor discovered to date. However, it is worth mentioning that in spite of the fact that the two-gap superconductivity was detected for Fe-based HTS belonging to different families (as *e.g.* 1111: [4, 6, 11, 12]; 122: [2, 5, 7, 9, 10, 13, 14]; 011: [15]) a systematic study of this phenomenon within one given family was not yet performed. In this paper we report on the evolution of two-gap behavior in the iron chalcogenide superconductor FeSe_{1-x} . The transition temperature was changed within the range $8.3 \lesssim T_c \lesssim 12.8$ K by applying an external pressure p between 0 and 0.84 GPa. At each particular pressure the superfluid density ρ_s was obtained from the in-plane magnetic penetration depth $\lambda_{ab}^{-2}(T) \propto \rho_s$ studied by means of muon-spin rotation, μSR . The analysis of $\lambda_{ab}^{-2}(T, p)$, performed by solving self consistently the gap equations derived within the two-gap scheme [16, 17], reveals that the main effect on $T_c(p)$

and $\lambda_{ab}^{-2}(T, p) \propto \rho_s(T, p)$ arises from the energy band(s) where the large superconducting gap, Δ_1 , develops. The zero-temperature values of Δ_1 , the contribution of this gap to the superfluid density $\lambda_{ab,1}^{-2}$, as well as the effective coupling constant Λ_{11} increase almost linearly with increasing T_c (increasing pressure). In contrast, the contribution of the small gap and thus Δ_2 , $\lambda_{ab,2}^{-2}$, and Λ_{22} , is practically pressure independent. Our results imply, therefore, that the transition temperature in FeSe_{1-x} is entirely determined by the intraband interaction within the band(s) where the dominant gap is opened.

The sample with the nominal composition $\text{FeSe}_{0.94}$ was prepared by solid state reaction similar to that described in Refs. 18, 19, 20. Powders of minimum purity 99.99% were mixed in appropriate ratios, pressed and sealed in a double-walled quartz ampoule. The sample was heated up to 700°C followed by annealing at 400°C [20]. The pressure was generated in a CuBe piston-cylinder type of cell especially designed to perform μSR experiments under pressure [21]. As a pressure transmitting medium 7373 Daphne oil was used. The pressure was measured in situ by monitoring the pressure shift of the superconducting transition temperature of Pb and/or In. The μSR experiments were carried out at the μE1 beam line, Paul Scherrer Institute, Switzerland. Zero-field (ZF) and transverse-field (TF) μSR experiments were performed at temperatures ranging from 0.24 to 50 K. For TF measurements the external magnetic field $\mu_0 H = 10$ mT was applied perpendicular to the muon-spin polarization. Typical counting statistics was $\sim 5 - 7 \cdot 10^6$ positron events for each data point.

The results of the ZF μSR experiments were previously reported in Ref. 22. It was shown that up to $p \simeq 0.8$ GPa the ZF response of $\text{FeSe}_{0.94}$ is determined by the contribution of the dilute Fe moments, in analogy with what was observed for ambient pressure measurements of $\text{FeSe}_{0.85}$ [15]. At $p = 0.84$ GPa static magnetism was found to occupy approximately 10% of the sample volume at $T \simeq T_c$ and it decreases down to $\sim 5\%$

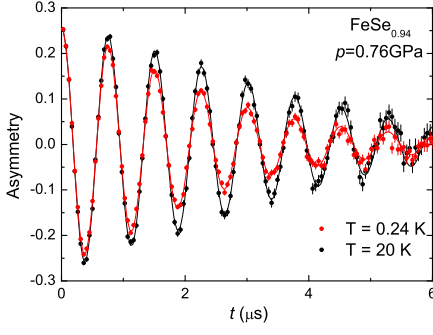


FIG. 1: (Color online) TF μ SR time-spectra ($\mu_0 H = 10$ mT) of $\text{FeSe}_{0.94}$ measured below ($T = 0.24$ K) and above ($T = 20$ K) the superconducting transition temperature ($T_c \simeq 13$ K) at $p = 0.76$ GPa. The stronger damping in the superconducting state is due to the formation of the vortex lattice.

at $T \simeq 0.25$ K [22].

Figure 1 shows the TF μ SR time-spectra measured at $p = 0.76$ GPa above ($T = 20$ K) and below ($T = 0.24$ K) the superconducting transition temperature ($T_c \simeq 13$ K). The stronger relaxation of the muon-spin polarization at 0.24 K relative to 20 K is due to the formation of the vortex lattice at $T < T_c$. The TF μ SR data were analyzed by using the functional form:

$$\begin{aligned} A(t) &= A_S(t) + A_{PS}(t) \\ &= A_{S,0} e^{-\Lambda t} e^{-\sigma_S^2 t^2/2} \cos(\gamma_\mu B_S t + \phi) \\ &\quad + A_{PS,0} e^{-\sigma_{PS}^2 t^2/2} \cos(\gamma_\mu B_{PS} t + \phi). \end{aligned} \quad (1)$$

Here the indexes S and PS denote the sample and the pressure cell, respectively, A_0 is the initial asymmetry, Λ is the exponential relaxation rate caused by the presence of diluted Fe moments [15], $\gamma_\mu = 2\pi \cdot 135.5$ MHz/T is the muon gyromagnetic ratio, B is the internal field, and ϕ is the initial phase of the muon-spin ensemble. The Gaussian relaxation rate, σ_{PS} , reflects the depolarization due to the nuclear magnetism of the pressure cell, while σ_S represents the depolarization in the sample arising from the nuclear moments and from the vortex lattice (see below). Each set of TF μ SR data taken at constant pressure was fitted simultaneously with $A_{S,0}$, $A_{PS,0}$, B_{PS} , σ_{PS} , Λ , and ϕ , as common parameters, and B_S and σ_S as individual parameters for each temperature point. The exponential relaxation rate Λ was assumed to be temperature independent in accordance with the results of ZF μ SR experiments [22].

In an anisotropic powder sample the magnetic penetration depth λ can be extracted from the Gaussian relaxation rate $\sigma_{sc}(T) = [\sigma_S^2(T) - \sigma_{nm}^2]^{1/2} \propto 1/\lambda^2(T)$, which probes the second moment of the magnetic field distribution in a superconductor in the mixed state [15, 23, 24]. Here σ_{nm} is the nuclear moment contribution measured at $T > T_c$. σ_{sc} can be converted into λ_{ab} via [15, 23]:

$$\sigma_{sc}^2/\gamma_\mu^2 = 0.00126 \Phi_0^2/\lambda_{ab}^4, \quad (2)$$

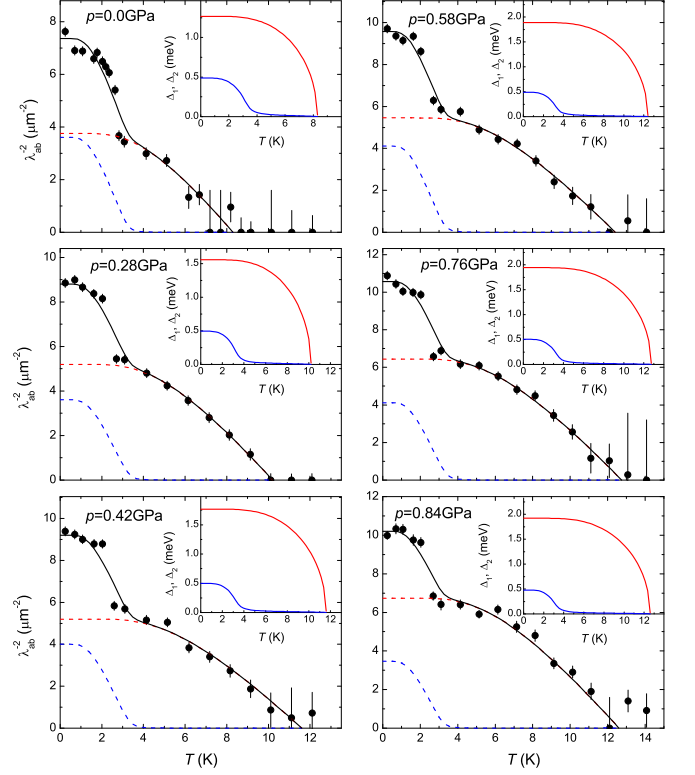


FIG. 2: (Color online) Temperature dependence of $\lambda_{ab}^{-2} \propto \rho_s$ of $\text{FeSe}_{0.94}$ measured at $p = 0.0, 0.28, 0.42, 0.58, 0.76$, and 0.84 GPa. The solid and the dashed lines are the theoretical curves obtained within the framework of the two-gap model described in the text. The insets show the temperature dependences of the large (Δ_1) and the small (Δ_2) gap.

where $\Phi_0 = 2.068 \cdot 10^{-15}$ Wb is the magnetic flux quantum. The measured $\lambda_{ab}^{-2}(T, p)$ of $\text{FeSe}_{0.94}$ at $p = 0.0, 0.28, 0.42, 0.58, 0.76$, and 0.84 GPa are shown in Fig. 2.

The experimental $\lambda_{ab}^{-2}(T)$ data were analyzed by using the two-gap model presented recently in Refs. 16, 17. Following [16], the coupled gap equations for a superconductor with k -dependent energy gaps: Δ_1, Δ_2 , intraband pairing potentials: V_{11}, V_{22} , and interband interaction potentials: V_{12}, V_{21} are determined as:

$$\begin{aligned} \Delta_i(k_i) &= \sum_{j=1,2} \sum_{k'_j} \frac{V_{i,j}(k_i, k'_j) \Delta_j(k'_j)}{2 \sqrt{E^2(k'_j) + \Delta_j^2(k'_j)}} \\ &\quad \times \tanh \frac{\sqrt{E^2(k'_j) + \Delta_j^2(k'_j)}}{2k_B T} \end{aligned} \quad (3)$$

Here $i = 1, 2$ is the band index and the sums are taken within the corresponding energy bands in k -space. Considering isotropic s -wave gaps [$\Delta_i(k_i) = \Delta_i$] and neglecting the momentum dependence of the band energies,

Eq. (3) reduces to:

$$\Delta_i = \sum_{j=1,2} \int_0^{\omega_{D_i}} \frac{N_j(0) V_{i,j} \Delta_j}{\sqrt{E^2 + \Delta_j^2}} \tanh \frac{\sqrt{E^2 + \Delta_j^2}}{2k_B T} dE, \quad (4)$$

Here ω_{D_i} is the phonon cutoff frequency (Debye frequency; note that these cutoffs are expected to be different for both bands $\omega_{D_1} \neq \omega_{D_2}$ [17]) and $N_i(0)$ is the partial density of states at the Fermi level. For convenience the sums were also converted into integrals [17]. A further simplification of Eq. (4) can be made by using the notation of the coupling constant $\Lambda_{ij} = N_i(0)V_{ij}$ introduced by Kogan *et al.* [25] and assuming similar cutoff frequencies for both bands ($\omega_{D_1} = \omega_{D_2} = \omega_D$):

$$\Delta_i = \sum_{j=1,2} \Lambda_{ij} \Delta_j \int_0^{\omega_D} \frac{1}{\sqrt{E^2 + \Delta_j^2}} \tanh \frac{\sqrt{E^2 + \Delta_j^2}}{2k_B T} dE. \quad (5)$$

The advantages to use the above equation rather than Eq. (4) is that (i) within the notation of Kogan *et al.* [25] $\lambda_{12} = \lambda_{21}$ and (ii) the total number of parameters needed to evaluate $\Delta_1(T)$ and $\Delta_2(T)$ reduces from 8 in case of Eq. (4) to 4 in case of Eq. (5).

With the known $\Delta_1(T)$ and $\Delta_2(T)$, λ_{ab}^{-2} can be obtained by decomposing it into two components $\lambda_{ab,1}^{-2}$ and $\lambda_{ab,2}^{-2}$ so that:

$$\lambda_{ab}^{-2}(T) = \lambda_{ab,1}^{-2}(T) + \lambda_{ab,2}^{-2}(T) \quad (6)$$

with [26]:

$$\frac{\lambda_{ab,i}^{-2}(T)}{\lambda_{ab,i}^{-2}(0)} = 1 + 2 \int_{\Delta_i(T)}^{\infty} \frac{\partial f}{\partial E} \frac{E}{\sqrt{E^2 - \Delta_i(T)^2}} dE.$$

Here $f = [1 + \exp(E/k_B T)]^{-1}$ is the Fermi function.

The analysis of $\lambda_{ab}^{-2}(T, p)$ by using the above described model was made by solely evaluating Λ_{22} , $\lambda_{ab,1}^{-2}(0)$, and $\lambda_{ab,2}^{-2}(0)$. The parameters Λ_{11} , Λ_{12} , and ω_D were taken as follows:

Λ_{12} : Our numerical analysis reveals that the step-like change of $\lambda_{ab}^{-2}(T)$ at $T \simeq 2.5$ K (see Fig. 2) requires the interband coupling constant Λ_{12} to be very small ($\Lambda_{12} \sim 10^{-3}$ or smaller). This implies that the band(s), where the large and the small superconducting energy gaps are open, become only weakly coupled. Note that a similarly small interband coupling constant was obtained by Kogan *et al.* for the superconductor V_3Si [25].

Λ_{11} : The fact that the interband coupling in $FeSe_{1-x}$ is weak thus suggests that the transition temperature T_c is mainly determined by the coupling within the band(s) where the large superconducting gap is opened. Assuming that the larger gap is Δ_1 , T_c is defined when $\Delta_1(T) = 0$, so that according to Eq. (5):

$$\Lambda_{11} \simeq \left[\int_0^{\omega_D} \frac{dE}{E} \tanh \frac{E}{2k_B T_c} \right]^{-1}. \quad (7)$$

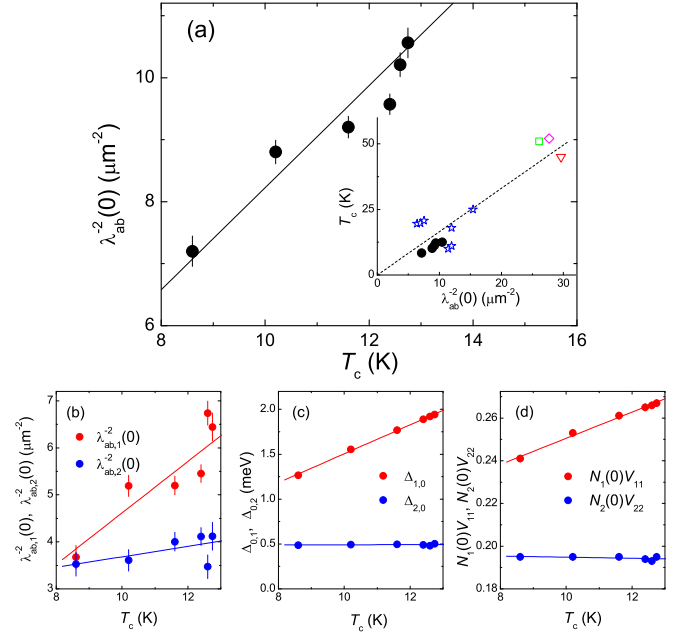


FIG. 3: (Color online) (a) Dependence of $\lambda_{ab}^{-2}(T = 0)$ on T_c . The inset is the “Uemura relation” for Fe-based HTS with some of data obtained to date (see Ref. 14, 15, 23, 30). (b), (c), and (d) Dependence of the parameters obtained in analysis of $\lambda_{ab}^{-2}(T, p)$ within the framework of two-gap model (see text for details).

With $T_c(p)$ measured independently (see Ref. [22]) Eq. (7) allows one to obtain the value of the intraband coupling constant Λ_{11} for each particular pressure.

ω_D : The ambient pressure value of the cutoff phonon frequency (Debye frequency) $\omega_D(p = 0) \simeq 40$ meV was taken from Ref. [27]. The increase of ω_D with increasing pressure was assumed to follow:

$$\omega_D(p) = \omega_D(0)(1 + \gamma p/B), \quad (8)$$

which is the consequence of the Grüneisen equation $\gamma = -d \ln \omega_D / d \ln V$ (γ is the Grüneisen parameter, $B \simeq 31$ GPa is the bulk modulus [19], and V is the sample volume). The Grüneisen parameter was assumed to be $\gamma \approx 1$ in analogy with Ref. 28. We should also emphasize that the parameters of the above described model are not very sensitive to the exact value of ω_D . As an example, the increase (decrease) of ω_D by a factor of 2 leads to a corresponding decrease(increase) of Λ_{11} obtained from Eq. (8) by $\simeq 15(18)\%$. This makes our assumption about using a similar cutoff phonon frequency for both bands [$\omega_{D_1} = \omega_{D_2}$, see Eq. (5)] to be rather reliable. Note that similar conclusion was also reached by Kogan *et al.* [29].

The parameters obtained from the analysis of $\lambda_{ab}^{-2}(T)$ by means of the model described above are summarized in Table I. The value of the interband coupling constant $\Lambda_{12} = 0.005$ was kept fixed. The red and the blue dashed lines in Fig. 2 correspond to the contribution of the large, $\lambda_{ab,1}^{-2}$, and the small, $\lambda_{ab,2}^{-2}$, superconducting gaps to the

TABLE I: Summary of the pressure studies of FeSe_{1-x}. The meaning of the parameters is: p – pressure; T_c – transition temperature; ω_D – Debye frequency; Λ_{12} – interband coupling constant, $\Lambda_{11}/\Lambda_{22}$, $\Delta_1(0)/\Delta_2(0)$, $\lambda_{ab,1}^{-2}(0)/\lambda_{ab,2}^{-2}(0)$ – intraband coupling constant, zero-temperature value of the gap, zero temperature value of superfluid density component within the band 1/2, respectively.

p (GPa)	T_c (K)	ω_D (meV)	Λ_{11}	Λ_{22}	Λ_{12}	$\Delta_1(0)$ (meV)	$\Delta_2(0)$ (meV)	$\lambda_{ab,1}^{-2}(0)$ (μm^{-2})	$\lambda_{ab,2}^{-2}(0)$ (μm^{-2})
0.00	8.3(1)	40.0	0.241(1)	0.195(1)	0.0005	1.27(1)	0.487(6)	3.67(25)	3.53(25)
0.28	10.2(1)	40.36	0.253(1)	0.195(1)	0.0005	1.56(1)	0.494(6)	5.19(23)	3.61(23)
0.42	11.6(1)	40.54	0.261(1)	0.195(1)	0.0005	1.77(1)	0.498(6)	5.20(20)	4.00(18)
0.58	12.4(1)	40.74	0.265(1)	0.194(1)	0.0005	1.89(1)	0.491(6)	5.45(19)	4.11(20)
0.76	12.8(1)	40.98	0.267(1)	0.195(1)	0.0005	1.94(1)	0.504(6)	6.44(29)	4.12(25)
0.84	12.6(1)	41.08	0.266(1)	0.193(1)	0.0005	1.92(1)	0.480(6)	6.74(29)	3.47(29)

total superfluid density, solid lines. The temperature dependences of the large, Δ_1 , and the small, Δ_2 , gaps are shown in the corresponding insets.

In order to check how the change of T_c affects the energy bands where the large and the small superconducting gaps are supposed to be open, we plot in Fig. 3 the parameters $\lambda_{ab}^{-2}(0)$, $\lambda_{ab,1}^{-2}(0)$, $\lambda_{ab,2}^{-2}(0)$, $\Delta_1(0)$, $\Delta_2(0)$, $\Lambda_{11} = N_1(0) V_{11}$, and $\Lambda_{22} = N_2(0) V_{22}$ as a function of the transition temperature T_c . From the obtained data the following conclusions can be drawn: (i) The zero-temperature superfluid density $\rho_s(0) \propto \lambda_{ab}^{-2}(0)$ increases with increasing T_c thus following the “Uemura” relation established recently for various Fe-based HTS, see Fig. 3 (a) and Refs. 14, 15, 23, 30. (ii) The electronic bands, where the large and the small gap are opened, are affected by the pressure quite differently. The increase of T_c with pressure leads to an almost linear increase of the superfluid density component $\lambda_{ab,1}^{-2}(0)$, the superconducting energy gap $\Delta_{1,0}$ as well as the effective coupling constant $\Lambda_{11} = N_1(0) V_{11}$. On the other hand, both $\Delta_{2,0}$ and $\Lambda_{22} = N_2(0) V_{22}$ stay almost constant, while $\lambda_{ab,2}^{-2}(0)$ increases with increasing T_c only slightly [see Figs. 3 (b), (c) and (e)]. Bearing in mind that the “large gap” and the “small gap” bands are only weakly coupled (the interband coupling constant Λ_{12} is estimated to be of the order of $5 \cdot 10^{-4}$ or less, see Table I) one may conclude that in the range of $0 \leq p \leq 0.84$ GPa the pressure effect on both T_c and λ_{ab}^{-2} is solely determined by the bands exhibiting the large superconducting gap.

To conclude, the superfluid density $\rho_s \propto \lambda_{ab}^{-2}$ was studied as a function of pressure and temperature in the superconductor FeSe_{1-x} by means of μSR . The analysis of $\rho_s(T)$ within a two-gap scheme reveals that the effect on both, T_c and $\rho_s(0)$, is entirely determined by the band(s) where the large superconducting gap develops. Our results suggests that for 011 family of Fe-based HTS the intraband interaction is most probably the leading pairing interaction determining the superconducting properties.

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